Electronic states of heavy element ions in nanophases

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Investigation of excited electronic states of heavy element ions has established directly the nanophase character of a new candidate nuclear waste form. Based on our research, this waste form encapsulates heavy metal ions in phosphate-rich nanophases that are embedded in glassy silica. Such encapsulation chemically fixes the sorbed metal ions and produces a robust, radiation-resistant material. The advantage of this waste form approach is reduction in nuclear waste volume because this material selectivity sorbs heavy element ions from nuclear waste solutions and, when subsequently heated in air, generates a nuclear waste form suitable for geologic disposal. The radiation-resistance of this material can be understood on the basis that its nanophase character causes most radiation damage within it to occur in its glassy silica component.

Isotopes of three heavy elements (curium, berkelium, and einsteinium) were incorporated into the waste form as trivalent ions. Nuclear decay of short-lived einsteinium-253 (20.47 day half-life) produced alpha particles and decay daughter berkelium ions. The resulting intense alpha radiolysis caused the curium ions in the waste form to emit the characteristic red luminescence seen in the self-luminescence photomicrograph in the highlight graphic. The decay daughter berkelium, due to nuclear recoil, traveled at most a few hundredths of a millionth of a meter. The nanophase character of the candidate waste form is established because that very slight movement caused decay daughter berkelium ions to be ejected from the einsteinium-containing heavy metal phosphate nanophases into the surrounding glassy silica. Due to such ejection, decay daughter berkelium ions exhibited a distinctly different luminescence emission spectrum and decay rate from that of berkelium ions that had been incorporated into the waste form by sorption from solution. Recoil-induced ejection was confirmed as nuclear decay of berkelium-249 generated californium ions that quenched berkelium ion luminescence from the originally sorbed berkelium ions but had little influence on the luminescence from decay daughter berkelium ions.

Initial evidence for the nanophase character of this material was originally obtained in small angle neutron scattering studies at the Intense Pulsed Neutron Source at Argonne National Laboratory. Development of this material as a nuclear waste form is being carried out as Nuclear Energy Research Initiative Project 99-0219. This candidate nuclear waste form is based on Diphosil, a material that was created with funding from the Division of Chemical Sciences by R. Chiarizia and coworkers in a collaboration between Argonne National Laboratory and the University of Tennessee. .Fundamental studies on the properties of heavy element ions in nanophases are being carried out as part our work for the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. The curium, berkelium, and einsteinium that we used in our studies were obtained from the Radiological Energy Development Center at Oak Ridge National Laboratory who produced these isotopes with funding from the Division of Chemical Sciences.

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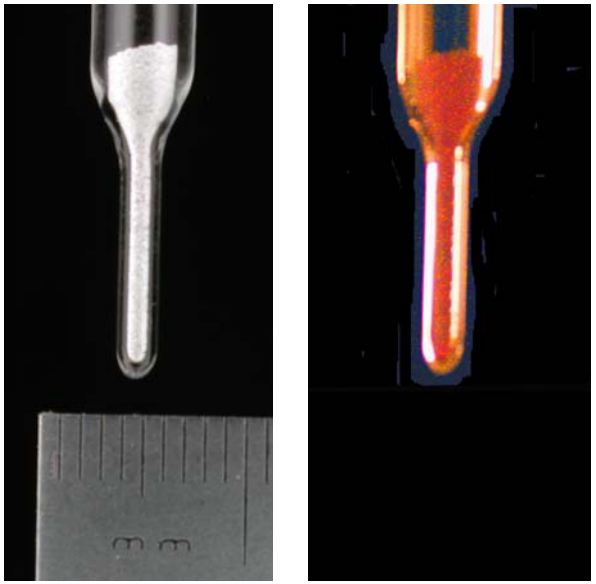
Investigation of excited electronic states of heavy element ions has established directly the nanophase character of a new candidate nuclear waste form that:

- **R**educes nuclear waste volume by selectively sorbing heavy element ions and serving as the basis of the final nuclear waste form.
- **E**ncapsulates heavy metal ions in phosphate-rich nanophases that are embedded in glassy silica.
- **A**chieves high radiation-resistance because most radiation damage occurs in its glassy silica component.



Reflected light (left) and self-luminescence (right) photomicrographs of a candidate nuclear waste form that contains curium, berkelium, and einsteinium isotopes. Alpha decay of einsteinium excites the characteristic red luminescence of electronically excited trivalent curium ions.

Development of this material as a nuclear waste form is being undertaken as part of a Nuclear Energy Research Initiative project. Fundamental studies on the properties of heavy element ions in nanophases are being carried out at Argonne National Laboratory under Contract No. W-31-109-ENG-38 with the Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Science, U. S. Department of Energy.